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(54) Title: PROCESS FOR EPOXIDIZING CRUDE POLYENES

(57) Abstract: A process for the epoxidation of a crude polyene, which process comprises the reaction of a mixture of unsaturated hydrocarbons which comprises one or more compounds selected from (especially polycyclic) polyenes and (co)oligomers thereof in a total concentration of from about 20 % to about 95 % by weight with aqueous hydrogen peroxide in the presence of a polyoxometallate-based catalytic system. This Abstract is not intended to define the invention disclosed in the specification, nor intended to limit the scope of the invention in any way.



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PROCESS FOR EPOXIDIZING CRUDE POLYENES

BACKGROUND OF THE INVENTION1. Field of the Invention

[0001] The present invention relates to a process for the epoxidation of crude polyenes, in particular, crude polycyclic polyenes such as, e.g., dicyclopentadiene (DCPD).

2. Discussion of Background Information

[0002] Numerous processes for the epoxidation of alkenes are known and it is possible to use a wide range of different reaction or catalyst systems. The epoxidation of alkenes in a homogeneous, liquid phase using organic hydroperoxides in the presence of catalysts based on molybdenum, tungsten or vanadium is carried out industrially. However, epoxide production is accompanied by the formation of equivalent or even greater amounts of alcohol corresponding to the hydroperoxide, and the need to utilize or recirculate this alcohol greatly restricts industrial use of this process. For this reason, alternative processes for a more direct epoxidation of alkenes are being developed to an increasing extent.

[0003] Another process for the oxidation of alkenes to epoxides is the reaction with hydrogen peroxide (H_2O_2). This process has been proposed for various epoxidation reactions because of, in particular, the positive aspect of the use of the oxidant in significantly reducing environmental pollution. Since the activity of hydrogen peroxide toward alkenes is only low, sometimes even completely absent, it is necessary to use activating agents, usually organic acids such as formic acid, acetic acid, etc., in organic solvents to facilitate the oxidation reaction. The acids form peracids in the reaction medium *in situ*, which peracids are the actual epoxidizing agent. These processes, too, do not appear to be particularly successful, because it is difficult to obtain the peracids and because of the instability of the epoxides in acidic media, as a result of which inconvenient process conditions are necessary. Moreover, the use of peracids also suffers from other disadvantages such as slow reaction, process safety issues, corrosion of equipment, cost of reagents and recovery and disposal of considerable quantities of carboxylic acid resulting from the reduction of the peracid.

[0004] For the epoxidation of polycyclic polyenes, for example, DCPD there are only a few industrial processes, which processes mainly use peracids (e.g., peracetic acid). It

would, therefore be desirable, to have available a process by which polyenes and in particular, polycyclic polyenes such as DCPD can be epoxidized by a simple, straightforward and inexpensive process which does not require the use of organic peracids and affords a high conversion of double bonds (in particular, a high epoxide ring content of the product). It would also be desirable for this process to be capable of using raw, unpurified and thus, relatively inexpensive polyene starting materials and nevertheless affording high double bond conversions. It would further be advantageous if the process afforded the epoxidation product in liquid form. Specifically, if the epoxidation product is a liquid the curing of the product to form epoxy resins is facilitated. Also, the process should advantageously afford no or only small amounts of heavy by-products.

SUMMARY OF THE INVENTION

[0005] The present invention provides a process for the epoxidation of a crude polyene. The process comprises reacting a mixture of unsaturated hydrocarbons which comprises one or more compounds selected from polyenes and oligomers (including co-oligomers) thereof in a total concentration of about 20 % to about 95 % by weight, based on monomeric polyene (i.e., treating the oligomers as if they were a plurality of monomeric polyenes), with aqueous hydrogen peroxide in the presence of a polyoxometallate-based catalytic system.

[0006] In one aspect, the mixture may comprise one or more compounds selected from polycyclic polyenes and oligomers and co-oligomers thereof in a total concentration of not more than about 94 % by weight, based on monomeric polycyclic polyene.

[0007] In another aspect, the one or more compounds may comprise at least one compound selected from dicyclopentadiene and oligomers and co-oligomers thereof.

[0008] In yet another aspect of the process, the one or more compounds may be present in a total concentration of at least about 70 % by weight.

[0009] In a still further aspect, the mixture may comprise (a) unsaturated hydrocarbons containing an average of from about 5 to about 55 carbon atoms per molecule and comprising not more than about 94 % by weight of a polycyclic polyene and/or (b) one or more oligomers and/or co-oligomers of hydrocarbon dienes having from 4 to about 18 carbon atoms, which (co)oligomers are based on at least about 6 % by weight of dienes which are different from a polycyclic polyene.

[0010] In another aspect of the process of the present invention, the mixture may comprise one or more unsaturated hydrocarbons selected from cyclopentene, cyclohexene, 2-methyl-2-butene, butadiene, isoprene, piperylene, cyclohexadiene, cyclopentadiene, dicyclopentadiene, methyl dicyclopentadiene, dimethyl dicyclopentadiene, limonene, dipentene, linear and cyclic dimers of piperylene, the norbornenes, norbornadiene, ethylidene norbornene, and dimers, codimers, oligomers and cooligomers of one or more of these unsaturated hydrocarbons.

[0011] In yet another aspect, the mixture may comprise from about 70 % to about 94 % by weight of dicyclopentadiene. The mixture may further comprise from about 6 % to about 30 % by weight of one or more C₉-C₁₂ dimers and/or codimers of one or more C₄-C₆ dienes and may additionally comprise (i) up to about 7 % by weight of one or more C₁₄-C₁₈ trimers and/or co-trimers of one or more C₄-C₆ dienes and/or (ii) up to about 10 % by weight of one or more compounds selected from aliphatic and cycloaliphatic diolefins.

[0012] In another aspect of the process, the mixture may comprise from about 20 % to about 70 % by weight of dicyclopentadiene, from about 1 % to about 10 % by weight of one or more C₉-C₁₂ dimers and/or co-dimers of one or more C₄-C₆ dienes, from 0 to about 10 % by weight of one or more oligomers of C₄-C₆ dienes and the balance to 100 % by weight of one or more compounds selected from C₄-C₆ alkanes, alkenes and dienes which are different from DCPD.

[0013] In another aspect of the present process, the mixture may comprise from about 30 % to about 70 % by weight of at least one C₄-C₆ diene and, optionally, up to about 10 % by weight of one or more C₉-C₁₂ dimers and/or co-dimers of one or more C₄-C₆ dienes, and the balance to 100 % by weight of one or more compounds selected from C₄-C₆ alkanes, alkenes and dienes which are different from a C₄-C₆ diene. By way of non-limiting example, the at least one C₄-C₆ diene may comprise piperylene and/or isoprene.

[0014] In another aspect of the present process, the epoxidation reaction may be carried out at a pH of less than about 5. For example, at least at the beginning (and preferably throughout the reaction) the reaction may be carried out at a pH of from about 3.5 to about 4.5. Specifically, the reaction may be carried out in the presence of a buffer. By way of non-limiting example, the buffer may comprise a mixture of (a) a tungstate compound; (b) a phosphoric acid compound; and (c) at least one inorganic base. The at least one inorganic

base may be selected from, e.g., alkali and alkaline earth metal compounds such as alkali and alkaline earth metal hydroxides.

[0015] In another aspect, the reaction may be carried out at a temperature of from not lower than about 10 °C, e.g., not lower than about 50 °C and not higher than about 100 °C, e.g., not higher than about 75 °C, for example at a temperature of not lower than about 60 °C and not higher than about 65 °C.

[0016] In yet another aspect of the process of the present invention, the polyoxometallate-based catalytic system may comprise at least one metal selected from Ti, Re, Mo, V, W and Mn. For example, the catalytic system may comprise W and in particular, a W/oxo/peroxo complex. A non-limiting example of such a complex is a compound of formula



wherein the radicals R independently represent a hydrocarbon group having from 1 to about 24 carbon atoms.

[0017] In a still further aspect of the process, the reaction may be carried out in the presence of a solvent. By way of non-limiting example, the solvent may comprise one or more hydrocarbon solvents and in particular, one or more solvents selected from halogenated hydrocarbons, cycloaliphatic hydrocarbons, and aromatic hydrocarbons.

[0018] In another aspect of the process, the hydrogen peroxide may be employed in a stoichiometric excess of up to about 20 % with respect to the theoretical amount required for a complete epoxidation of all double bonds present in the mixture.

[0019] In another aspect, the hydrogen peroxide may be added to the mixture at a temperature of from about 50 °C to about 70 °C over a period of at least about 30 minutes, e.g., at least about 60 minutes to not more than about 180 minutes, e.g., not more than about 120 minutes.

[0020] In another aspect, at least about 80 %, e.g., at least about 85 %, at least about 90 %, at least about 95 % or even at least about 98 % of all epoxidizable double bonds present in the mixture may be converted by the process.

[0021] In yet another aspect, the process may afford a non-solid (and preferably a liquid) reaction product.

[0022] The present invention also provides a process of preparing a curable epoxy resin composition. The process comprises combining an epoxidation product that is obtainable by the process of the present invention as set forth above, including the various aspects thereof, and a curing agent. By way of non-limiting example, the curing agent may comprise one or more of methyl tetrahydrophthalic anhydride (MTHPA), hexahydrophthalic anhydride (HHPA), and methyl hexahydrophthalic anhydride (MHHPA).

[0023] The present invention also provides a curable epoxy resin composition which comprises an epoxidation product that is obtainable by the process of the present invention as set forth above (including the various aspects thereof) and a curing agent.

[0024] The present invention further provides a cured epoxy resin which is obtainable by curing an epoxidation product that is obtainable by the process of the present invention as set forth above (including the various aspects thereof) with a curing agent.

DETAILED DESCRIPTION OF THE PRESENT INVENTION

[0025] The particulars shown herein are by way of example and for purposes of illustrative discussion of the embodiments of the present invention only and are presented in the cause of providing what is believed to be the most useful and readily understood description of the principles and conceptual aspects of the present invention. In this regard, no attempt is made to show embodiments of the present invention in more detail than is necessary for the fundamental understanding of the present invention, the description making apparent to those skilled in the art how the several forms of the present invention may be embodied in practice.

[0026] The process of the present invention is less geographically dependent because it only requires a reaction section and a separation section, without the infrastructure and expenditure related to the production of a peracid such as peracetic acid (the use whereof involves a complex process, a highly corrosive medium, high energy consumption, etc.). Moreover, the process is less expensive and affords better yields than the peracid process. The process also can be used with non-purified raw materials. In particular, for DCPD the starting material may advantageously contain, for example, about 70 % to about 94 % by weight of DCDP. By using this kind of stream one may obtain a final epoxide product that is liquid at room temperature, which is an advantage, for example, if the product is to be cured to form epoxy resins. The epoxide products of the present invention are useful as

intermediates in the production of other products, for example, products which are suitable for use in the fields of coatings, laminates, composites, encapsulations, and molding compositions.

[0027] The process of the present invention is suitable for the epoxidation of various crude polyenes and polyene mixtures, respectively, and is capable of affording a product which has a high epoxide ring content. Non-limiting examples of feed streams (starting materials) which can advantageously be used in the process of the present invention include:

(a) unsaturated hydrocarbons which contain an average of from about 6 to about 55 carbon atoms per molecule and contain not more than about 94 % by weight of DCPD;

(b) oligomers and/or co-oligomers of hydrocarbon dienes, which dienes comprise 4 to about 18 carbon atoms and comprise at least about 6 % by weight of dienes which are different from DCPD; and

(c) mixtures of (a) and (b) in any ratio.

[0028] Non-limiting examples of suitable unsaturated hydrocarbons which can be employed in the process of the present invention either in a crude state or in a purified state and either alone or in any combination of two or more thereof, include cyclopentene, cyclohexene, 2-methyl-2-butene, butadiene, isoprene, piperylene, cyclohexadiene, cyclopentadiene, dicyclopentadiene, methyl dicyclopentadiene, dimethyl dicyclopentadiene, limonene, dipentene, linear and cyclic dimers of piperylene, norbornenes, norbornadiene, ethylidene norbornene, (co)dimers and (co)oligomers of one or more of these unsaturated hydrocarbons, and the like.

[0029] An example of a particularly suitable mixture of unsaturated hydrocarbons for use in the present invention is a DCPD concentrate which comprises from about 70 % to about 94 % by weight of DCPD, from about 6 % to about 30 % by weight of C₉₋₁₂ dimers and/or codimers of C₄₋₆ dienes such as, for example, cyclopentadiene-isoprene, cyclopentadiene-piperylene, cyclopentadiene-methyl cyclopentadiene and/or dimers of isoprene, piperylene, methyl cyclopentadiene and the like; optionally, up to about 7 % by weight of C₁₄₋₁₈ trimers and/or cotrimers of C₄₋₆ dienes; and, optionally, up to about 10 % by weight of aliphatic diolefins such as, for example, piperylene, isoprene, 1,5-hexadiene, and/or cyclic olefins such as, for example, cyclopentadiene, methyl cyclopentadiene, cyclopentene and the like. Methods for the preparation of such DCDP concentrates and more detailed descriptions

thereof can be found in, e.g., U.S. Patent Nos. 3,557,239 to Gebhart et al. and 4,167,542 to Nelson, the entire disclosures whereof are incorporated by reference herein.

[0030] Another example of a particularly suitable mixture for use in the process of the present invention is a crude DCPD stream which comprises from about 20 % by weight to about 70 % by weight of DCPD, from about 1 % to about 10 % by weight of dimers and/or codimers of C_{4-6} dienes as set forth above, optionally, up to about 10 % by weight of oligomers of C_{4-6} dienes and the balance to 100 % of C_{4-6} alkanes, alkenes and dienes different from DCPD.

[0031] Yet another example of a mixture which can advantageously be employed in the process of the present invention is a crude piperylene and/or isoprene stream which comprises from about 30 % to about 70 % of piperylene and/or isoprene, optionally, up to 10 % by weight of C_{9-12} dimers and/or codimers of C_{4-6} dienes (as set forth above) and the balance to 100 % of C_{4-6} alkanes, alkenes and dienes which are different from piperylene and/or isoprene.

[0032] The hydrogen peroxide used in the process of the present invention is preferably employed in the form of an aqueous solution. Aqueous hydrogen peroxide is commercially available in a wide range of concentrations, for example, from about 5 wt % to about 70 wt %. A preferred aqueous hydrogen peroxide solution for use in the present invention has a concentration of about 20 % to about 35 % by weight, e.g., about 30 % by weight. The amount of hydrogen peroxide used in the present invention preferably is at least an about stoichiometric amount with respect to the epoxidizable double bonds which are present in the starting material. Preferably, an up to about 20 % stoichiometric excess with respect to the theoretical amount required for a complete epoxidation of the double bonds of the starting material is employed.

[0033] In the epoxidation reaction mixture of the present invention the hydrogen peroxide is generally employed in a total amount of from about 1 wt % to about 20 wt %, preferably from about 4 wt % to about 16 wt %, and more preferably from about 8 wt % to about 12 wt %, based on the total weight of the reaction mixture.

[0034] The epoxidation reaction of the present invention is carried out in the presence of a polyoxometallate-based catalyst. The catalyst preferably contains a transition metal. Compounds which are useful as catalyst for the epoxidation reaction of the present invention are preferably selected from homogeneous and heterogeneous catalysts which are

based on metals such as, e.g., Ti, Re, Mo, V, W and Mn. Preferably, the catalyst comprises one or more W/oxo/peroxo complexes. An example of a particularly preferred catalyst for use in the process of the present invention is an amphiphilic quaternary ammonium peroxotungstophosphate. By way of non-limiting example, the catalyst used in the present invention may comprise one or more of the catalysts described in Journal of Organic Chemistry (1988), 53, pp. 1553 – 1557; U.S. Patent Nos. 4,562,276; 4,595,671 and 5,274,140; and European Patent Applications EP 1 170 291 A1 and 0 493 778 A1; the entire disclosures of all of these documents are expressly incorporated by reference herein.

[0035] The catalyst for use in the process of the present invention may be in the form of a solid, for example, a solid prepared in accordance with the process described in the above Journal of Organic Chemistry article; the catalyst may further be in the form of a liquid composition, e.g., a liquid composition as described in EP 1 170 291 A1. The catalytic system described in EP 1 170 291 A1 includes a composition comprising a tungsten compound, a quaternary onium salt and a mineral acid.

[0036] The tungsten compounds which may be used for the epoxidation catalyst for use in the present invention may be selected, for example, from tungsten-containing inorganic acids and salts of these acids. Non-limiting specific examples of tungsten-containing acids and salts thereof include tungstic acid (wolframic acid) and salts thereof, for example, sodium tungstate, potassium tungstate, lithium tungstate, ammonium tungstate; dodecatungstates, for example, sodium dodecatungstate, potassium dodecatungstate and ammonium dodecatungstate; heteropolyacids and salts thereof, for example, phosphotungstic acid, sodium phosphotungstate, silicotungstic acid, sodium silicotungstate, phosphovanadotungstic acid; and phosphomolybdotungstic acid. Preferably, the tungsten-containing acids and salts thereof comprise at least one of tungstic acid, sodium tungstate, potassium tungstate, and phosphotungstic acid. The above tungsten compounds may be employed alone or as a mixture of two or more thereof.

[0037] Preferably, the tungsten compound(s) for the catalyst of the process of the present invention is/are employed in an amount of from about 0.0007 wt % to about 5 wt %, more preferably from about 0.002 wt % to about 3 wt %, calculated as tungsten, based on the total weight of the reaction mixture.

[0038] Non-limiting examples of the quaternary onium salts which may be used for the epoxidation catalyst of the process of the present invention include quaternary ammonium

halides, for example, trioctylmethyl ammonium chloride, tridecylmethyl ammonium chloride, trioctylmethyl ammonium bromide, benzyldimethyltetradecyl ammonium chloride, benzyltriethyl ammonium chloride, dimethyldidodecyl ammonium chloride, benzyltributyl ammonium chloride, benzyltributyl ammonium iodide and phenyltrimethyl ammonium chloride; quaternary ammonium hydrogen sulfates such as, for example, trioctylmethyl ammonium hydrogen sulfate; quaternary ammonium perchlorates, for example, trioctylmethyl ammonium perchlorate; quaternary ammonium dihydrogen phosphates, for example, trioctylmethyl ammonium dihydrogen phosphate; quaternary ammonium nitrates, for example, trioctylmethyl ammonium nitrate; quaternary ammonium hydrosilicofluorates, for example, trioctylmethyl ammonium hydrosilicofluorate; and quaternary ammonium acetates, for example, trioctylmethyl ammonium acetate. Quaternary ammonium halides, especially trioctylmethyl ammonium chloride and tridecylmethyl ammonium chloride, alone or in combination, are particularly preferred.

[0039] The content of the quaternary onium salt in the epoxidation catalyst may, for example, be from about 0.0003 wt % to about 4 wt %, and preferably from about 0.003 wt % to about 2.5 wt %, based on the total weight of the reaction mixture.

[0040] Non-limiting examples of mineral acids which may be used for the epoxidation catalyst of the process of the present invention include phosphoric acid, sulfuric acid, hydrochloric acid, perchloric acid, hexafluorosilicic acid, nitric acid and tetrafluorosilicic acid. Preferably, phosphoric acid and sulfuric acid, more preferably phosphoric acid, are employed for the epoxidation catalyst. The above-mentioned mineral acids may be employed alone or as a mixture of two or more thereof. The content of the mineral acid in the epoxidation catalyst may, for example, be from about 0.001 wt % to about 5 wt %, and preferably from about 0.005 wt % to about 3 wt %, based on the total weight of the reaction mixture.

[0041] More preferably, a catalyst for use in the process of the present invention may be a catalyst as described in J. Org. Chem. 1988, 53, 1553-1557 and illustrated by the formula:



wherein the radicals R independently represent hydrocarbon (e.g., alkyl) groups having from 1 to about 24 carbon atoms. Non-limiting examples of the radicals R in the above formula include alkyl groups such as CH₃, C₆H₁₃, C₈H₁₇, C₁₆H₃₃, and C₁₈H₃₇. Usually, the radicals R include at least one, and preferably at least two hydrocarbon (e.g., alkyl) groups

which comprise at least about 6 carbon atoms. Also, preferably the total number of carbon atoms in the four radicals R is at least about 24. Non-limiting examples of the group R_4N^+ in the above catalyst formula include (a) $[(C_6H_{13})_4N]^+$, (b) $[(C_8H_{17})_3NCH_3]^+$ and (c) $\{[C_{18}H_{37} (76\%) + C_{16}H_{33} (24\%)]_2N(CH_3)_2\}^+$.

[0042] A preferred catalyst for the epoxidation reaction of the present process is a catalyst which comprises a cation of type (c) above and is usually employed in an amount of from about 0.1 wt % to about 1.5 wt %, preferably from about 0.2 wt % to about 1.2 wt %, and more preferably from about 0.4 wt % to about 1 wt %, based on the total weight of the reaction mixture.

[0043] The epoxidation of the present invention is preferably conducted by controlling the pH of the reaction medium such as, for example, by adding a buffer solution to the reaction medium. Controlling the pH can result in a significant yield improvement and/or a significant reduction in the formation of heavy by-products.

[0044] Preferably, a buffering agent is added to the reaction mixture to maintain the reaction mixture at a pH of less than about 5 during the epoxidation reaction. In particular, for certain polyenes whose epoxides tend to be subject to an acid-catalyzed hydrolysis the reaction is best performed at a pH of the aqueous phase which is suitable to substantially prevent a ring opening reaction of the corresponding epoxides.

[0045] In a preferred aspect, the buffering agent may comprise a mixture of components which include the following three materials: (a) a tungstate for assisting in maintaining the activity of the catalyst; (b) a mineral acid, preferably, phosphoric acid also for assisting in maintaining the activity of the catalyst; and (c) an alkali and/or alkaline earth metal salt and/or an ammonium compound for maintaining the pH of the reaction mixture.

[0046] Component (a) of the buffering agent may be any of the tungsten compounds described above with reference to the epoxidation catalyst. Component (a) preferably includes one or more of $Na_2WO_4 \cdot 2H_2O$, K_2WO_4 , $[NH_4]_2WO_4$. Preferably, the molar ratio component (a) : epoxidation catalyst is from about 0:1 to about 5:1.

[0047] Component (b) of the buffering agent may be any of the mineral acids described above with reference to the catalyst. Preferably, component (b) comprises or essentially consists of H_3PO_4 . Preferably, the molar ratio component (b) : epoxidation catalyst is from about 0:1 to about 30:1.

[0048] Component (c) of the buffering agent may, for example, be an aqueous alkali solution. For example, the aqueous alkali solution may comprise one or more compounds

selected from ammonia and the basic compounds of alkali metals and alkaline earth metals. The aqueous alkali solution has a pH value of higher than about 7, preferably at least about 8, more preferably at least about 10, still more preferably at least about 11. [0049] Non-limiting examples of the basic compounds which may be present in the aqueous alkali solution include hydroxides, carbonates, bicarbonates and sulfites of alkali metals (e.g., Li, Na and K) and hydroxides, carbonates, bicarbonates and sulfites of alkaline earth metals (e.g., Mg, Ca and Ba). Preferably, the hydroxides, carbonates, bicarbonates and sulfites of alkali metals are employed, and more preferably the hydroxides of alkali metals are employed. Specific and non-limiting examples of the basic compounds for use in component (c) include ammonium hydroxide, potassium hydroxide, sodium hydroxide, magnesium hydroxide, barium hydroxide, calcium hydroxide, potassium carbonate, sodium carbonate, magnesium carbonate, calcium carbonate, potassium bicarbonate, sodium bicarbonate, potassium sulfite and sodium sulfite. Preferably, ammonium hydroxide, sodium hydroxide, potassium hydroxide and sodium sulfite, more preferably ammonium hydroxide, sodium hydroxide and potassium hydroxide are employed in the buffering agent. The basic compounds may be employed alone or in a mixture of two or more thereof.

[0050] Component (c) is preferably used in an amount which is sufficient to result in a pH of the aqueous phase of from about 3.5 to about 4.5, for example, of about 4. Component (c) is preferably used in a molar ratio of component (c) : epoxidation catalyst of from about 5:1 to about 20:1

[0051] Optionally, the epoxidation reaction of the present invention may advantageously be carried out in an inert solvent. Non-limiting examples of suitable inert solvents that may be useful in the present process include halogenated hydrocarbons, cycloaliphatic hydrocarbons, aromatic hydrocarbons and mixtures thereof. Non-limiting specific examples thereof include benzene, toluene and aromatic solvents, dichloromethane, dichloromethane and chlorinated solvents, hexane, aliphatic and cycloaliphatic alkenes, and mixtures thereof.

[0052] When a solvent is used in the process of the present invention the amount thereof based on the total weight of the reaction mixture is generally from about 0.0001 wt % to about 90 wt %, preferably from about 0.5 wt % to about 60 wt %, and more preferably from about 30 wt % to about 50 wt %.

[0053] In the epoxidation reaction in accordance with the process of the present invention the organic solvent may be contained as a reaction medium in the reaction system. There is no limitation with respect to the type of the organic solvent as long as the organic solvent is

not detrimental to the epoxidation reaction. The organic solvent for the reaction medium includes aliphatic halogenated hydrocarbons, for example, chloroform, dichloroethane, and dichloromethane; aliphatic non-halogenated hydrocarbons, for example, cyclohexane and n-heptane; and aromatic hydrocarbons, for example, benzene, toluene and xylene. The above-mentioned organic solvents may be employed alone or as a mixture of two or more thereof.

[0054] When an organic solvent is employed, the weight of the organic solvent preferably is not higher than about 20 times, more preferably not higher than about 10 times the total weight of the mixture which comprises the epoxidizable compounds.

[0055] The epoxide compounds of the present invention are produced by the epoxidation of the olefinic double bonds contained in the starting material. Generally, the epoxidation reaction may be conducted over a wide range of reaction conditions. For example, the temperature range of the reaction may generally be from about 10 °C to about 100 °C, and preferably from about 50 °C to about 75 °C, e.g. from about 60 °C to about 65 °C.

[0056] The required reaction time depends, *inter alia*, on the nature and on the quantity of the catalyst, on the solvent and on the polyenes used in the process. In general, the reaction time can be from minutes to hours for completing the reaction. Preferably, the epoxidation reaction time ranges from about 1 hour to about 20 hours, most preferably from about 4 hours to about 6 hours. For example, especially if the starting material comprises a significant percentage of DCPD (e.g., from about 40 % to about 50 % by weight), particularly preferred reaction conditions include adding the aqueous hydrogen peroxide (preferably an about 30 % by weight solution) over a period of about 1 to about 2 hours, followed by a "digestion time" of from about 3 to about 5 hours. As set forth above, a particularly preferred reaction temperature is from about 60 °C to about 65 °C and the pH is preferably kept at a value of about 4 by using, for example, a buffer comprising $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$, phosphoric acid (e.g., about 85 % phosphoric acid) and NaOH (e.g., about 10 % aqueous NaOH).

[0057] The pressure range for the epoxidation reaction is generally from vacuum to about 30 atmospheres (atm), most preferably from about 1 atm to about 5 atm. Usually, ambient (atmospheric) pressure will be employed.

[0058] The order of addition of the starting materials is not critical and any order can be used. Preferably, the mixing is carried out in the following order: first, the organic solvent (e.g., toluene) and the catalyst (as a solid, dissolved in a solvent, or in the form of separate catalyst components) are added to the reactor; thereafter the crude polyene is added to the

reactor; and then the H_2O_2 /buffering agent is added to the reaction mixture. The reaction mixture is thoroughly mixed under preferably vigorous mixing conditions.

[0059] The reaction may optionally be carried out under an inert gas atmosphere such as, e.g., a nitrogen atmosphere.

[0060] Preferably, the epoxidation reaction is carried out in a biphasic liquid system which comprises a liquid phase which comprises the epoxidizable starting material and another liquid phase which comprises the aqueous hydrogen peroxide solution, phase-separated from each other. For example, the epoxidation reaction may be carried out by vigorously mixing the epoxidizable starting material, an aqueous hydrogen peroxide solution and a catalyst comprising a transition metal compound with each other in an atmosphere that may essentially consist of an inert gas such as for example, nitrogen gas, and by heating the resultant mixture under ambient atmospheric pressure or increased pressure, while thoroughly agitating the mixture. There is no limitation with respect to the reaction temperature. As mentioned above, the reaction temperature usually is from about 10 °C to about 100 °C, and preferably from about 50 °C to about 75 °C. After completion of the reaction, the product may be recovered by any convenient means known to those skilled in art, such as, for example, distillation or extraction. The catalyst may also be separated from the reaction by well known means in the art.

[0061] As an illustration of one embodiment of the process of the present invention, the process may be conducted using a reactor fitted with a stirrer, a heat-controlling system and a reflux coolant. Pre-established quantities and ratios of the reactants (H_2O_2 and the polyene containing mixture in a solvent) are introduced into the reactor. The catalyst and buffering agent are also introduced into the reactor in the desired quantities. Under vigorous stirring, the heterogeneous mixture is brought to the reaction temperature for the desired time. At the end of the reaction time, the reaction mixture separates into two phases. After cooling down of the reaction mixture, the epoxidized product and the reactants may be separated by conventional means and methods such as distillation or extraction. Isolating the epoxidized product from the resulting reaction mixture can be carried out by any technique, examples of which are well known to those skilled in the art, and the present invention is not limited to any particular method for isolating the reaction product from the reaction mixture.

[0062] The process of the present invention generally provides high yields, for example preferably higher than 90 %; and a high selectivity, preferably higher than 90 % of product, which makes the process attractive for the industrial application of the process.

[0063] The epoxidation process of the present invention provides a product with advantageous properties, particularly when using the epoxide product for various end uses. The epoxide products prepared by the process of the present invention are useful as intermediates in the production of products which are suitable for use in the fields of, e.g., coatings, inks, adhesives, laminates, composites, potting, encapsulating, and molding compositions.

[0064] By way of non-limiting example, the epoxidized product prepared by the process of the present invention may be useful as a component of a curable epoxy resin composition wherein the epoxy product is reacted with a curing agent, together with other typical ingredients, for curing the resin composition to provide thermosets for the various end uses mentioned above. The curable epoxy resin composition containing the epoxidized product made by the process of the present invention can also contain one or more additional epoxy compounds, solvents, catalysts, plasticizers, fillers, pigments and/or any other commonly used additives for the application being employed.

[0065] The epoxy product prepared according to the present invention can be used alone or in combination with other epoxy compounds to manufacture cured epoxy resins by reacting the epoxy materials with well known epoxy curing agents. Such curing agents include, for example, amine-curing agents such as dicyandiamide, diaminodiphenylmethane and diaminodiphenylsulfone; anhydrides such as hexahydroxyphthalic anhydride and styrene-maleic anhydride copolymers; imidazoles; and phenolic curing agents such as phenol novolac resins; and mixtures thereof. Such curing agents can be added to a resin composition immediately before curing, or can be included in the composition from the beginning if the curing agents are latent. The amount of the curing agent used may normally range from about 0.3 to about 1.5 equivalent per epoxy equivalent of the epoxy components, and preferably from about 0.5 to about 1.1 equivalent per epoxy equivalent of the epoxy components.

[0066] Non-limiting specific examples of curing agents include tetrahydrophthalic anhydride (THPA), methyl tetrahydrophthalic anhydride (MTHPA), hexahydrophthalic anhydride (HHPA), methyl hexahydrophthalic anhydride (MHHPA), nadic methyl anhydride (NMA), polyazelaic polyanhydride, succinic anhydride, maleic anhydride,

phthalic anhydride, and the like, and mixtures of anhydrides. Preferably, the curing agent used in the present invention includes one or more of methyl tetrahydrophthalic anhydride (MTHPA), hexahydrophthalic anhydride (HHPA), and methyl hexahydrophthalic anhydride (MHHPA).

[0067] A typical epoxy resin composition containing the epoxide product prepared by the process of the present invention may also comprise, as an optional component, catalysts for catalyzing the reaction of the epoxy product and the curing agent. Non-limiting examples of the suitable catalysts are imidazoles such as 2-methylimidazole; 2-phenyl imidazole and 2-ethyl-4-methyl imidazole; tertiary amines such as triethylamine, tripropylamine and tributylamine; phosphonium salts such as ethyltriphenylphosphonium chloride, ethyltriphenylphosphonium bromide and ethyltriphenylphosphonium acetate; and ammonium salts such as benzyltrimethylammonium chloride and benzyltrimethylammonium hydroxide, and mixtures thereof.

[0068] Cationic photoinitiators, which release an acid when exposed to ultra violet (UV) or electron-beam radiation, may be also used as catalysts for catalyzing reactions of the epoxy product of the present invention, including, for example, homopolymerization and reactions of the epoxy product and other epoxy compounds, oxetanes, and hydroxyls for example. Examples of suitable cationic photoinitiators include aryl sulfonium salts and aryl iodonium salts containing non-nucleophilic anions which are capable of curing epoxy resins when exposed to UV or electron-beam radiation. The amount of the catalysts generally ranges from about 0.001 wt % to about 2 wt %, and preferably from about 0.01 wt % to about 0.5 wt %, based on the total weight of the reaction mixture. The amount of cationic photoinitiator may vary from about 0.001 wt % to about 20 wt %, and preferably from about 0.1 wt % to about 10 wt %.

[0069] Non-limiting specific examples of catalysts include benzyldimethylamine (BDMA), 1,4-diazabicyclo[2.2.2]octane (DABCO), tertiary amines, imidazole and derivatives of imidazole, 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU), chlorine, benzyl triethyl ammonium chloride, ethyl triphenyl phosphonium iodide, benzyl triphenyl phosphonium bromide, tetraethyl ammonium bromide, Mark DBVIII from Witco, stannous octoate, zinc octoate and mixtures of two or more of these catalysts. Preferably, the catalyst includes one or more of benzyldimethylamine, diazabicyclo[2.2.2]octane (DABCO), imidazole and derivatives of imidazole.

[0070] Non-limiting examples of cationic photoinitiators include mixed triaryl sulfonium hexafluorophosphate salts, for example CYRACURE UVI-6992, and mixed triaryl sulfonium hexafluoroantimonate salts, for example CYRACURE UVI-6976, products of The Dow Chemical Company; and mixed diaryl iodonium hexafluorophosphate salts, for example IRGACURE 250, a product of Ciba Specialty Chemical Company.

[0071] Non-limiting examples of thermally cured compositions wherein the epoxy product of the present invention may be used as a component of a resin composition in significant concentrations include insulators for instrument transformers, switch gear, and bushings; medium voltage power line insulators; light-emitting diode (LED) encapsulants; coatings for wire used to make electrical motors; potting compounds for capacitors; high voltage electrical arresters; filament windings for pressurized rocket fuel tanks and aerospace composites; and crosslinkers for automotive topcoatings.

[0072] Non-limiting examples of UV cured compositions wherein the epoxy product of the present invention may be used as a major ingredient include coatings on plastic tubes (e.g. tubes used for personal care products); coatings on steel can ends (e.g. vegetable cans); coatings on steel containers (e.g. biscuit tins and aerosol cans); white base coatings used on laminated steel beverage cans; UV inks on film and foil used for packaging; rapid prototyping; and electronic coatings.

[0073] The epoxy product of the present invention may also be used as an acid scavenger composition, for example, wherein the product is used as an additive present at less than about 5 % of a total composition, including, for example, an acid scavenger for an organophosphate hydraulic fluid; an acid scavenger for polyvinyl chloride (PVC) siding for homes; an acid scavenger for brominated flame retardant composition; and an acid scavenger used in chemical manufacturing processes.

[0074] Preferred applications for the curable epoxy resin composition and the cured resin include coatings, inks, potting, and encapsulating compositions.

[0075] The present invention will be further illustrated by the following non-limiting examples.

Example 1 (reference example)

[0076] Toluene, catalyst and DCPD in the amounts set forth in Table I below are charged in a 1 liter (L) continuous stirred tube reactor (CSTR) jacketed glass reactor equipped with a mechanical stirrer, thermocouple and reflux condenser. The catalyst is of the approximate formula $(R_4N)_3^+ \{PO_4[W(O)_2(O_2)_2]_4\}^{3-}$ wherein $R_4N^+ = [C_{18}H_{37} \text{ (76\%)} + C_{16}H_{33}$

(24%) $]$ $_2\text{N}(\text{CH}_3)_2]^+$. The temperature of the reaction is regulated at a temperature of from 60 °C (for initial 2.5 hours) to about 65 °C (for the final 2.5 hours, including cooling time) by setting the temperature of the heating oil which is continuously pumped to the reactor jacket. In view of the exothermicity of the reaction between DCPD and H_2O_2 a water coil is mounted internally in the reactor in order to control the temperature of the reaction and maintain the temperature fixed at the desired value. The reaction is conducted at atmospheric pressure (about 1 bar) and the magnetic stirrer speed is set at 600 rpm to ensure satisfactory contact between the organic and aqueous phases.

[0077] A mixture of a solution of hydrogen peroxide (concentration about 30 wt %) and a buffering agent, as set forth in Table I below, is placed in a calibrated glass cylinder mounted on an HPLC pump suction side. The glass cylinder is connected to a balance in order to control its flow rate and the time needed to feed the solution into the reactor (about 60 minutes).

Table I

		MW	mmoles	g
Hydrogen Peroxide	H_2O_2	34	1524.62	51.84
H_2O_2 solution	31.8%	-	-	162.96
$\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$		329.86	7.95	2.62
H_3PO_4		98	39.77	3.90
H_3PO_4 (85 %)				4.59
NaOH 10%				14.58
Total				184.75

		MW	mmoles	g
Catalyst		2758	2.65	7.31
DCPD		132	1325.76	175.00
Toluene				236.76
Water				0.00
Total				419.08

Total Reactor Batch	603.83
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[0078] The hydrogen peroxide/buffering mixture solution is fed with an HPLC pump for 1 hour; after feeding is stopped (after 60 minutes) and an amount corresponding to a molar ratio of 2.3/1 with respect to the DCPD has been added (15 % H₂O₂ excess over stoichiometric ratio), the reaction is allowed to continue for 4 hours. At the end of the this time period the resulting mixture is cooled down to room temperature (about 25°C) in 5-10 minutes and discharged into a 1.5 liter separatory funnel, leaving two separate phases - an organic phase and an aqueous phase. The two phases are then analyzed and characterized using the analytical methods set forth in Table II below.

Table II

Phase	Analysis	Method
Reaction Mixture/Organic Phase	Organic Phase Composition	Gas Chromotography (GC)
Reaction Mixture/Aqueous Phase	Residual H ₂ O ₂ content	Iodometric Titration
	pH Determination	Potentiometric

[0079] The product obtained in toluene solution is a mixture of 2,4-methano-2H-indeno[1,2-b:5,6-b']bisoxirene, octahydro- (9CI) ("DCPD diepoxy") and monoepoxide of 4,7-methano-1H-indene, 3a,4,7,7a-tetrahydro- (9CI) ("DCPD monoepoxy"). The results of the reaction are summarized in Table III below.

Table III

COMPONENT	Concentration (wt %)
DCPD	0.00
DCPD Monoepoxy	1.00
DCPD Diepoxy	46.00
Toluene	53.00
	(Mol-%)
DCPD Conversion	100.00
(Monoepoxy + Diepoxy) Yield	98.00

Example 2

[0080] Example 2 is carried out by repeating Example 1 except that a crude (impure) DCPD product (30 % wt % of other components) is used as starting material. Details of the reaction mixture are set forth in Table III below.

Table III

		MW	mmoles	g
Hydrogen Peroxide	H ₂ O ₂	34	1524.62	51.84
H₂O₂ solution	31.8%	-	-	162.96
Na₂WO₄*2H₂O		329.86	7.95	2.62
H₃PO₄		98	39.77	3.90
H₃PO₄ (85 %)				4.59
NaOH 10%				14.58
Total				184.75

	MW	mmoles	g
Catalyst	2758	2.65	7.31
DCPD	132	1325.76	175.00
Others			52.50
Toluene			236.76
Water			0.00
Total			471.58

Total Reactor Batch	656.33
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The results of the reaction are summarized in Table IV below.

Table IV

COMPONENT	Concentration (wt %)
DCPD	0.00
Others	11.00
DCPD Monoepoxy	2.00
DCPD Diepoxy	40.00
Toluene	47.00
	(Mol-%)
DCPD Conversion	100.00
(Monoepoxy + Diepoxy) Yield	96.00

[0081] It is noted that the foregoing examples have been provided merely for the purpose of explanation and are in no way to be construed as limiting of the present invention. While the present invention has been described with reference to exemplary embodiments, it is understood that the words which have been used herein are words of description and illustration, rather than words of limitation. Changes may be made, within the purview of the appended claims, as presently stated and as amended, without departing from the scope and spirit of the present invention in its aspects. Although the present invention has been

described herein with reference to particular means, materials and embodiments, the present invention is not intended to be limited to the particulars disclosed herein; rather, the present invention extends to all functionally equivalent structures, methods and uses, such as are within the scope of the appended claims.

WHAT IS CLAIMED IS:

1. A process for the epoxidation of a crude polyene, wherein the process comprises reacting a mixture of unsaturated hydrocarbons which comprises one or more compounds selected from polyenes and (co)oligomers thereof in a total concentration of from about 20 % to about 95 % by weight, based on monomeric polyene, with aqueous hydrogen peroxide in the presence of a polyoxometallate-based catalytic system.
2. The process of claim 1, wherein the mixture comprises one or more compounds selected from polycyclic polyenes and (co)oligomers thereof in a total concentration of not more than about 94 % by weight, based on monomeric polycyclic polyene.
3. The process of any one of claims 1 and 2, wherein the one or more compounds comprise at least one compound selected from dicyclopentadiene and (co)oligomers thereof.
4. The process of any one of claims 1 to 3, wherein the one or more compounds are present in a total concentration of at least about 70 % by weight.
5. The process of any one of claims 1 to 4, wherein the mixture comprises at least one of (a) unsaturated hydrocarbons containing an average of from about 5 to about 55 carbon atoms per molecule and comprising not more than about 94 % by weight of a polycyclic polyene, and (b) one or more (co)oligomers of hydrocarbon dienes having from 4 to about 18 carbon atoms, which (co)oligomers are based on at least about 6 % by weight of dienes which are different from a polycyclic polyene.
6. The process of any one of claims 1 to 5 wherein the mixture comprises one or more unsaturated hydrocarbons selected from cyclopentene, cyclohexene, 2-methyl-2-butene, butadiene, isoprene, piperylene, cyclohexadiene, cyclopentadiene, dicyclopentadiene, methyldicyclopentadiene, dimethyldicyclopentadiene, limonene, dipentene, linear and cyclic dimers of piperylene, norbornenes, norbornadiene, ethylidene norbornene, and

dimers, codimers, oligomers and cooligomers of one or more of these unsaturated hydrocarbons.

7. The process of any one of claims 1 to 6, wherein the mixture comprises from about 70 % to about 94 % by weight of dicyclopentadiene.

8. The process of claim 7, wherein the mixture further comprises from about 6 % to about 30 % by weight of one or more C₉-C₁₂ (co)dimers of one or more C₄-C₆ dienes.

9. The process of claim 8, wherein the mixture further comprises at least one of (i) up to about 7 % by weight of one or more C₁₄-C₁₈ (co)trimers of one or more C₄-C₆ dienes, and (ii) up to about 10 % by weight of one or more compounds selected from aliphatic and cycloaliphatic diolefins.

10. The process of any one of claims 1 to 6, wherein the mixture comprises from about 20 % to about 70 % by weight of dicyclopentadiene, from about 1 % to about 10 % by weight of one or more C₉-C₁₂ (co)dimers of one or more C₄-C₆ dienes, from 0 to about 10 % by weight of one or more oligomers of C₄-C₆ dienes and a balance to 100 % by weight of one or more compounds selected from C₄-C₆ alkanes, alkenes and dienes different from dicyclopentadiene.

11. The process of claim 1, wherein the mixture comprises from about 30 % to about 70 % by weight of at least one C₄-C₆ diene, from 0 % to about 10 % by weight of one or more C₉-C₁₂ (co)dimers of one or more C₄-C₆ dienes and a balance to 100 % by weight of one or more compounds selected from C₄-C₆ alkanes, alkenes and dienes different from a C₄-C₆ diene.

12. The process of claim 11, wherein the at least one C₄-C₆ diene comprises at least one of piperylene and isoprene.

13. The process of any one of claims 1 to 12, wherein the reaction is carried out at an initial pH of less than about 5.

14. The process of any one of claims 1 to 13, wherein the reaction is carried out at an initial pH of from about 3.5 to about 4.5.

15. The process of any one of claims 1 to 14, wherein the reaction is carried out in the presence of a buffer.

16. The process of claim 15, wherein the buffer comprises a mixture of (a) a tungstate compound; (b) a phosphoric acid compound; and (c) an inorganic base.

17. The process of claim 16, wherein the inorganic base is selected from alkali and alkaline earth metal compounds.

18. The process of any one of claims 1 to 17, wherein the reaction is carried out at a temperature of from about 10 °C to about 100 °C.

19. The process of any one of claims 1 to 18, wherein the reaction is carried out at a temperature of from about 60 °C to about 65 °C.

20. The process of any one of claims 1 to 19, wherein the polyoxometallate-based catalytic system comprises at least one metal selected from Ti, Re, Mo, V, W and Mn.

21. The process of any one of claims 1 to 20, wherein the polyoxometallate-based catalytic system comprises a W/oxo/peroxo complex.

22. The process of any one of claims 1 to 21, wherein the polyoxometallate-based catalytic system comprises a compound of formula



wherein the radicals R independently represent a hydrocarbon group having from 1 to about 24 carbon atoms.

23. The process of any one of claims 1 to 22, wherein the reaction is carried out in the presence of a solvent.

24. The process of claim 23, wherein the solvent comprises one or more compounds selected from halogenated hydrocarbons, cycloaliphatic hydrocarbons, and aromatic hydrocarbons.

25. The process of any one of claims 1 to 24, wherein the hydrogen peroxide is employed in a stoichiometric excess of up to about 20 % with respect to a theoretical amount required for a complete epoxidation of all double bonds present in the mixture.

26. The process of any one of claims 1 to 25, wherein the hydrogen peroxide is added to the mixture at a temperature of from about 50 °C to about 70 °C over a period of from about 30 minutes to about 120 minutes.

27. The process of any one of claims 1 to 26, wherein at least about 80 % of all double bonds are converted.

28. The process of any one of claims 1 to 27, wherein at least about 95 % of all double bonds are converted.

29. The process of any one of claims 1 to 28, wherein the process affords a non-solid mixture of epoxy compounds.

30. An epoxidation product obtainable by the process of any one of claims 1 to 29.

31. A process of preparing a curable epoxy resin composition, wherein the process comprises combining an epoxidation product obtainable by the process of any one of claims 1 to 29 and a curing agent.

32. A curable epoxy resin composition comprising an epoxidation product obtainable by the process of any one of claims 1 to 29 and a curing agent.

33. A cured epoxy resin obtainable by curing an epoxidation product obtainable by the process of any one of claims 1 to 29 with a curing agent.